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SOME REGULARITIES OF DISTRIBUTION AND MIGRATION
OF RADIOACTIVE ELEMENTS IN SOIL COVERING

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When studying the radioactivity of the soil and plant cover, one should bear in mind that at present it is due to the existence of natural and artificial radioelements which differ not only by their physico-chemical properties, but by their origin as well. The latter is of particularly great significance in studying the behaviour of radioactive elements coming from the atmosphere.

This work is aimed at obtaining an over-all pattern of the average content and distribution of natural radioelements in types of soil in the various climatic zones of the USSR, as well as at revealing the regularities of distribution and migration of artificial radioelements (in the case of Sr^{90}), depending on the sum total of the natural conditions.

1. Natural radioactivity of soil covering

The basic components of natural radioactivity of the soil is the radiation of the radioisotopes of the uranium and thorium families, as well as K^{40} , whose energy amounts to about 98% of the total sum of natural radiation.

The samples of soil to determine natural radioactivity were taken by genetic horizons according to the methods generally accepted in soil science.

The samples were used to determine radium and thorium by the radiochemical (emanation) method. Uranium was determined by the luminescent method, and potassium by the method of flame photometry. The average content of radioelements in some

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basic types of soil in the European part of the USSR by genetic horizons is given in Table 1.

The content of radioelements in the investigated soil of the USSR ranged from $4 \cdot 10^{-6}\%$ to $16 \cdot 10^{-4}\%$ for thorium (for an air-dry sample), and from $1 \cdot 10^{-12}\%$ to $1.7 \cdot 10^{-10}\%$ for radium. Direct determination of **uranium** content in some samples of the soil has shown that the radioactive equilibrium between uranium and radium is deranged but slightly ^{/1/}. The content of uranium, calculated from the content of radium, ranges from $3 \cdot 10^{-6}\%$ to $5.1 \cdot 10^{-4}\%$.

The ratio of Th: Ra is within the range from $0.9 \cdot 10^6$ to $14.4 \cdot 10^6$, which corresponds to Th: U ranging from 0.3 to 4.8.

The content of radioelements in most soils is determined by their content in the soil-forming rock. For instance, the concentration of Th, Ra and U, and the ratio Th : U in the soil covering clay and sand is close to their content in the parent rock. At the same time some kinds of soil, as for example those formed on products of erosion of carbonate rock, and peat deposits sharply differ from the **underlying rock by the** concentration of radioelements.

The highest radioactivity is encountered in sod-meadow alluvial loamy soil of river flood-plains (Th = $15 \cdot 10^{-4}\%$, Ra = $1.6 \cdot 10^{-10}\%$), and the least in sand soil and particularly in peat deposits of high moors.

The content of radioelements in peat deposits of high **moors** (Th = $4-8 \cdot 10^{-6}\%$; Ra = $0.8-2.2 \cdot 10^{-12}\%$) is lower by an order than that in peat deposits of **low-land bogs** (Th = $2-9 \cdot 10^{-5}\%$; Ra = $1-2.3 \cdot 10^{-11}\%$).

The distribution and ratio of the radioelements in the soil genetic horizons reflects the soil-forming processes, including the part played by organic matter.

The differentiation of the radioelements is **feebly marked** by their profile in many kinds of soil (black earth, grey forest soil and some kinds of sod-podzolic soil) (Figs. 1 and 2).

The upper horizons of soil **/A₀; A₁/** (bedding and humus)

are appreciably impoverished by Ra and Th (Fig. 2 P - 82, Fig. 3 - 123). The enrichment of the upper horizons and diminution of the concentration of radioelements is typical of many flood-plain alluvial kinds of soil.

An accumulation of radioelements has, as a rule, been recorded in illuvial /B/ (Fig. 3) and gley /G/ horizons of various types of soil.

The accumulation of radioelements in illuvial and gley horizons appears to be related to the fine clayey fraction of the soil. This is attested by the parallelism of the Ra and Th distribution curves, and by the mechanical fraction which is less than 0.01 mm and 0.001 mm.

In well-developed soil, where the thickness of the humus horizon A is as great as 20 cm, agricultural cultivation does not affect the distribution of natural radioisotopes by the soil profile. This is borne out by the Ra and Th distribution charts by the soil profile of arable and virgin black earth (Fig. 1), sod-podzolic (Fig. 2) and sod-calcareous soil (Figs. 3 and 4).

In the case of very thin soil bedded with poorly permeable dense bedrock, ploughing changes fundamentally the initial pattern of distribution of radioelements in similar virgin soil. This has been well demonstrated by the instance of the so-called alvars of the Estonian Republic, humus-calcareous soil bedded with agglutinated Silurian limestone at a depth from 10 to 20 cm (Fig. 5).

Organic matter plays a special part in the migration of chemical elements in the soil covering. Like other microelements, radioisotopes can penetrate into organisms and accumulate there while the latter are alive. After death, organic matter is capable of actively absorbing radioelements and accumulating them under dynamic conditions if they are delivered and added together with water solutions.

If radioelements had accumulated in considerable quantities by organisms during their life,^u the upper horizons of every type of soil, filled with humus, would have been noted

a higher concentration of Ra, Th and U. Actually the litter-fall /A₀/ and the sod and humus horizons /A₁/ are noted for a relative drop in the Ra and Th contents as compared with illuvial /B/ and parent /C/ horizons.

The peat deposits of high moors and of low-land bogs consist almost entirely of biomass, but the concentration of radioelements even in the ash residue does not exceed the content in other kinds of soil.

For instance, the content of Th in the ash of peat deposits is twice or three times lower than in other kinds of soil. The content of Ra in the ash residue of high moors is, as a rule, much lower than in loamy soil, and in the ash of low-land bog peat deposits it is the same as in the loamy soil of river flood-plains /2/.

Hence, organic matter is not the principal factor in the accumulation of Ra and Th in the soil.

Active migration of natural radioelements does not seem to occur in the soil, which is supported by the following regularities known by this time.

1. The total content of natural radioelements in the soil is inherited from parent soil-forming rock.

2. There is a slight differentiation of natural radioelements by genetic horizons in podzolic, grey forest, chernozem and some other kinds of soil.

3. There is no change in the redistribution for two or three years of radioelements in thin soil on dense bedrock, arising as a result of ploughing (in fields sown with perennial grass).

2. Artificial radioactivity of soil covering

As is well known, radioactive fission products, when falling to the ground, are distributed on it unevenly, this being attended with a so-called latitudinal pattern of distribution. Proceeding from numerous data on the content of radioactive strontium in the soil covering of many countries ^{3,4},

including the Soviet Union, it has been established that the maximum content of this isotope is encountered at 40-50° latitude North, despite the rather great spread within one and the same latitude zone (Fig. 6).

The recorded non-uniformity in the distribution of radioactive strontium and, in all probability, of other artificial radioactive elements is basically due to the geographic position of the observation point, the regularities of the circulation of the atmosphere and the exchange of the mass of air between the troposphere and the stratosphere, as well as to the sum total of the natural conditionsⁱⁿ which the radioelements have got.

The major part of the radioelements that fall from the atmosphere are retained in the upper 15 cm of soil covering⁵⁻⁹, but even within this horizon (0-5 and 5-15 cm) there is a different distribution. This is explained by the fact that radioactive isotopes, similar to ordinary chemical elements, are subjected to processes of redistribution, i.e. concentration and dispersion, under the influence of natural factors (the structure of the earth's surface, peculiarities of the soil and plant cover, climatic and hydrological conditions, etc.).

The research was carried out in the forest and steppe zones of the Soviet Union with the application of the comparative geographic method which permits to establish the peculiarities of distribution of some or other element at sections of Earth surface conjugated by runoff. Simultaneously with studying the nature of distribution of strontium-90, investigation were conducted on the behaviour of stable strontium, calcium and other chemical elements.

The effect of natural factors on the nature of distribution and redistribution of radioactive elements in the soil covering is most markedly manifested at the period when there is a minimum of their reaching the earth's surface. From this standpoint, 1961, before the beginning of a new series of new nuclear explosions, was the most suitable year to study the

regularities of distribution of radioactive fission products in soil covering.

It has been found in the case of strontium-90 that its content in the soil covering of the Soviet Union practically did not change in 1960 as compared with 1959, which testifies to the fact that a mobile equilibrium set in 1960, i.e. the amount of radioactive strontium that came to the earth's surface corresponded to that which was displaced from the surface horizons of the soil as a result of its migration along the soil profile and penetration into plants. A similar phenomenon was also observed by other researchers ⁴.

As noted above, even level virgin open areas with a minimum runoff considerably differ by the content of strontium-90 in the upper surface horizon within the same latitude zone.

The irregular nature of distribution in the soil is manifested to a still greater extent when studying not only under placor conditions, but under other conditions as well. Of substantial importance in this case is also the type of the soil covering.

If the types of soil occurring under placor conditions are to be aligned in a series according to a decrease in the average content of strontium -90: chernozem > grey forest > > podzolic, quite a different pattern of redistribution can be observed within one and the same soil and climatic zone, depending on the natural factors. This is illustrated by the data in Table II.

Particularly sharp contrasts are manifested in the forest zone, on areas conjugated by the runoff and even lying at a small distance from one another (within 200 - 400 m). The lowest content of strontium-90 is typical of soddy-podzolic sand soil, and the highest, of gley soil of bogged up flood-plains and peat soil of watershed swamps. In the latter cases the content of this isotope is twice or three times higher than in the zone of chernozem and higher by an

order than its average content in soddy-podzolic soil located on level placor areas with a minimum runoff.

The above pattern of redistribution of radioactive strontium in various natural zones is not casual and is due to natural conditions.

For instance, soddy-podzolic sand soil is characterized by a low content of humus and exchange bases, an acid reaction, a low content of total calcium and stable strontium, and a good capacity for filtration. All this makes for radioactive strontium, when it partly becomes fixed in a shallow, low-humus horizon A_1 , to migrate together with the surface and intrasoil runoff toward conjugately located areas of bogged up flood-plains where it wedges in together with subsoil water and gets a firm hold in humus- gley and soddy-gley soil. Typical of the latter is a high content of humus and exchange bases, a less acid reaction and a smaller capacity for filtration. Typical of bogged up flood-plains is also the concentration of other chemical elements (calcium, strontium, manganese, iron, etc.).

The higher content of strontium-90 in the peat soil of watershed swamps, which are not, as a rule, points of concentration of other elements, is due, in all probability, both to the direction of the surface and the intrasoil runoff and the retaining capacity of sphagnum moss.

The more uniform distribution of radioactive strontium in the chernozem of the steppe zone is due to the greater homogeneity of the soil covering, the high content of humus and of exchange bases, the greater depth of the humus horizon, and to the peculiarity of the water regime.

Simultaneously with studying the horizontal migration of strontium-90 owing to the surface and intrasoil runoff, we also investigated its vertical migration, depending on the genetic structure of the soil.

The data in Fig. 7 indicate that there is a dependence of the distribution of strontium-90 in various types of the

soil on the peculiarities of the humus horizon, the nature of the humus and its depth.

In soil with a thin humus horizon (podzolic sand), up to 80-90% of strontium-90 are concentrated in a coarse humus layer from one to two cm deep, lying between a fresh, loose bedding and podzolic horizons. This is most likely due both to the formation of the layer at the period of maximum fall-out in 1959 (its age is about two years) and its genetic peculiarities.

In soddy-gley periterrace soil with a deeper and well-pronounced silty-humus horizon and a sharp transition to a gley horizon, strontium-90 is likewise concentrated in the humus horizon, spreading more or less uniformly throughout its depth. It is practically not to be found then in the upper 0-3 cm of the loose bedding of grass stalks, and in the gley horizon.

Strontium-90 is distributed more uniformly in peat soil than in soddy-podzolic soil and is to be found at a greater depth; it is the upper 0-5 cm layer, however, that contains its maximum. Such a pattern of distribution of radioactive strontium is due, on the one hand, to the peculiarities of location of this soil in the area where intrasoil water wedges out and, on the other hand, to the peculiarities of their organic matter.

In typical loamy chernozem soil, radioactive strontium penetrates to a great depth; its maximum content has been recorded, however, in the upper part of the humus horizon (3-8 cm), lying under the loose grass bedding. A slight increase in the content of this isotope at a depth of 20-30 cm is to be found in leached black chernozem with a distribution of a similar nature as in typical chernozem soil. This corresponds to the increase in the content of stable strontium and calcium, due to the processes of illuviation.

In arable soddy-podzolic and chernozem soil, the average content of strontium-90 coincides, in the main, with that in corresponding virgin watershed soil. The fluctuations

observed in the arable soil of one and the same zone, located at a small distance from one another (from 5 m to 1.5 km) is due to the degree of cultivation, the nature of tilling and the peculiarities of the crops farmed there.

Radioactive strontium is distributed more uniformly within an arable horizon than in virgin soil; the subtile horizon is characterized by a decrease in the content of this isotope.

Table 1

Average content of radioelements in the basic types of soil in the USSR as percentage of an air-dry weight (according to V.I. Baranov, K.G. Kunasheva and N.G. Morozova)

Item N°	Soil and climatic zone and type of soil	Genetic horizon	Th 10 ⁻⁴ %	Ra 10 ⁻¹⁰ %	U 10 ⁻⁴ %	K %	Th:U
1	2	3	4	5	6	7	8
<u>Forest zone</u>							
1.	Soddy-calcareous (loamy), with limestone bed- ding	A ₁ B C D	7.7 10.0 4.1 2.0	1.0 1.3 0.6 0.25	3.0 3.9 1.8 0.75	1.50 1.80 0.40 0.39	2.6 2.56 2.28 2.66
2.	Soddy-podzolic, loamy	A ₀ A ₁ A ₂ B ₁ B ₂ C	6.7 6.5 9.2 9.5 8.9	0.65 0.77 1.00 0.95 0.82	1.95 2.31 3.00 2.85 2.46	1.40 1.60 1.70 1.70 1.58	3.44 2.71 3.07 3.33 3.62
3.	Soddy-podzolic, sandy	A ₀ A ₁ A ₂ B ₁ B ₂ C	2.0 2.0 2.0 2.0 3.0	0.20 0.30 0.25 0.30 0.20	0.60 0.90 0.75 0.90 0.60		3.34 2.22 2.67 2.22 5.00
4.	Soddy-meadow, flood-plain alluvial, loamy	A ₁ (0-10) A ₁ (10-75) C	13.0 16.4 10.1	1.40 1.74 1.15	4.20 5.22 3.45	1.72 2.34 2.64	3.1 3.15 2.93
5.	Soddy-gley, heavy loamy and clayey	A ₀ A B ₁ g B ₂ g Cg	4.5 3.0 7.5 6.5	1.15 1.40 1.25 0.90	3.45 4.20 3.75 2.70	2.1 2.9 3.1 2.6	1.3 1.9 2.0 2.4

Table 1 (continued)

1	2	3	4	5	6	7	8
6.	Peat deposits of low-land bogs	A _T	0.33	0.15	0.45		0.73
		A _T /G	2.5	0.45	1.35		1.85
7.	Peat deposits of high moors	A _T	0.06	0.014	0.042		1.43
	<u>Forest-stepped zone</u>						
8.	Grey forest, loamy	A ₁	8.7	0.90	2.70	2.30	3.22
		B ₁	9.5	1.10	3.30	2.55	2.88
		B ₂	9.4	1.00	3.00	2.50	3.13
		C	10.0	1.05	3.15	1.25	3.17
9.	Heavy loamy chernozem	A ₁	8.0	0.85	2.55	1.80	3.13
		B ₁	9.0	1.07	3.21	1.50	2.80
		B ₂	9.3	0.89	2.67	1.80	3.48
		C	10.4	1.00	3.00	1.74	3.47
	<u>Zone of dry steppes</u>						
10.	Chestnut, slightly alkaline, slightly loamy	A ₁	5.0	0.60	1.80	1.06	2.78
		B ₁	8.0	0.70	2.10	1.80	3.80
		B _k	6.0	0.80	2.40	1.58	2.50
		C	6.0	0.70	2.10	1.41	2.86

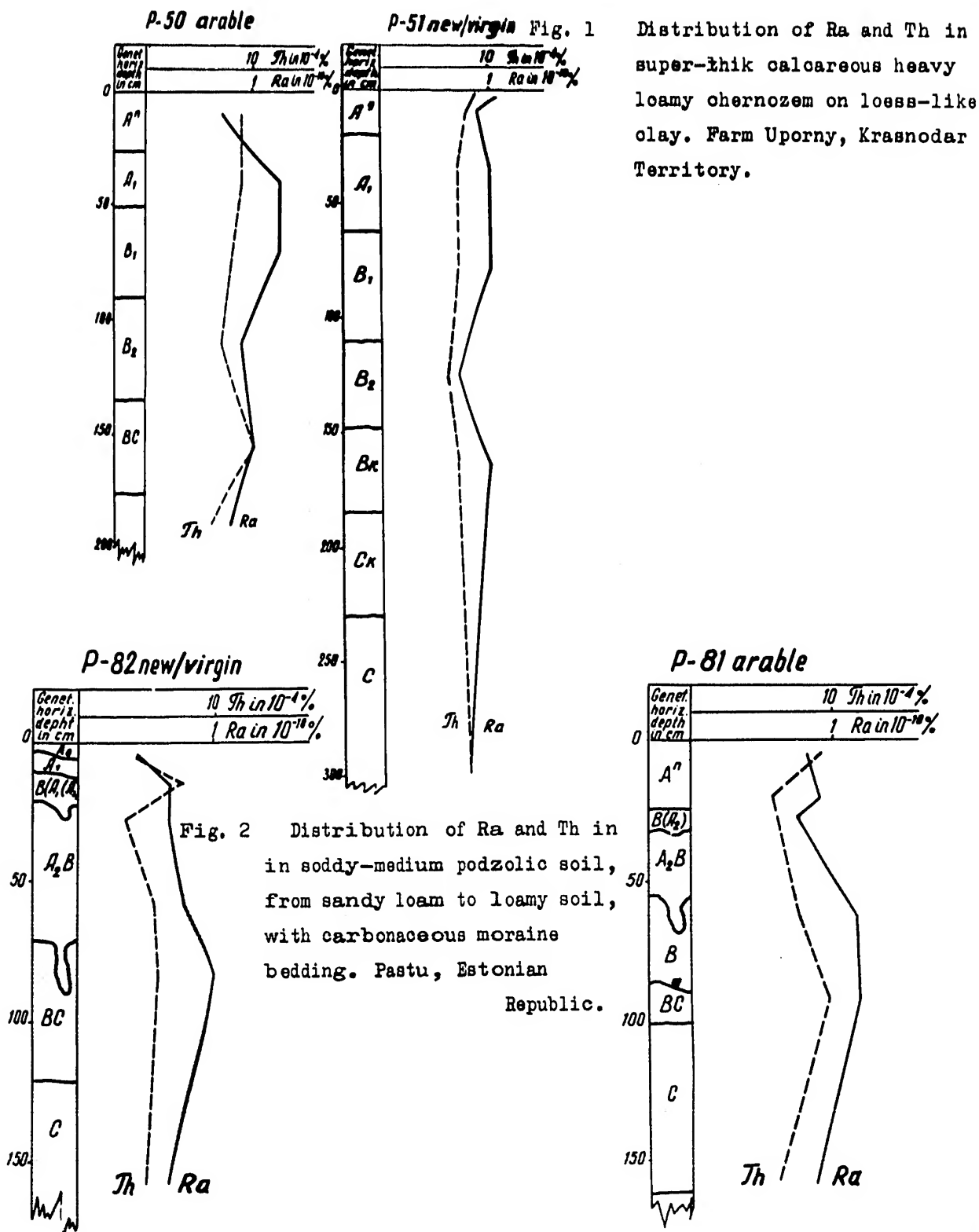
Table II

Content of strontium-90 (10^{-9} curie/m²) in a 0-20 cm
layer of various types of soil

Soil	Content of strontium-90
1. Podzolic, sandy, under forest	8.0-13.9
2. Light brown, slightly loamy	23.0
3. Peaty-humus-gley, sandy loam, under forest	7.6-23.6
4. Humus-gley, sandy loam of bogged up depressions	21.6-23.9
5. Peaty-ferruginous-gley of a bogged up depression	29.8
6. Thin peat, of a watershed transitional swamp	28.6
7. Soddy-gley, sandy loam of a bogged up sedge flood-plain	75.3
8. Soddy-gley, ferruginous, of alder periterrace	94.1
9. Podzolic, loamy, arable	5.8-20.5
10. Podzolic, sandy loam, arable, much tilled	11.1-16.6
11. Grey, forest, loamy, arable	12.8-27.1
12. Typical thick loamy chernozem	22.0-38.2
13. Chernozem podzolized, loamy	28.2-31.3
14. Thick feably leached loamy chernozem	23.9-32.2
15. Typical thick loamy arable chernozem	20.7-32.2

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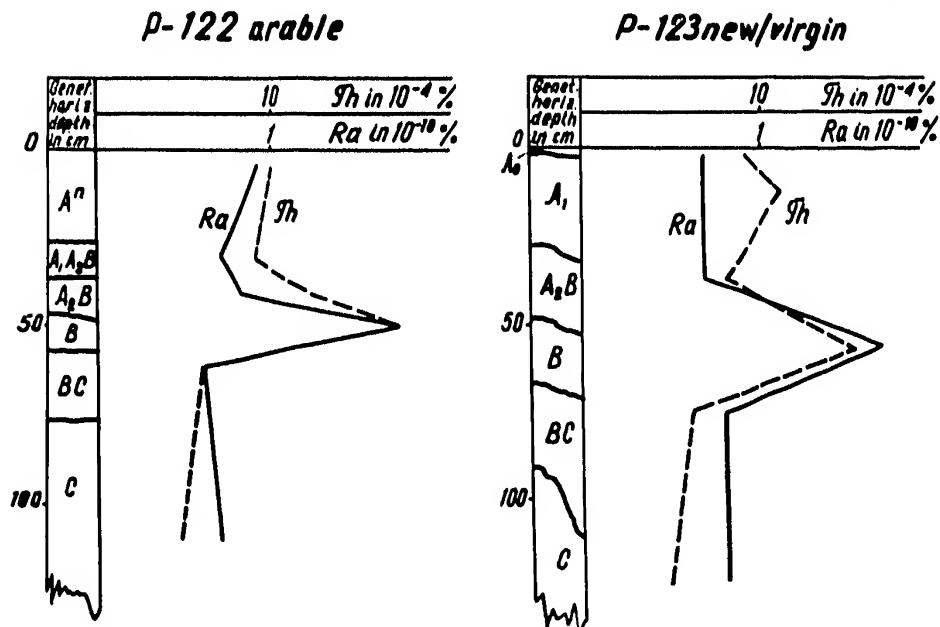


Fig. 3 Distribution of Ra and Th in soddy-calcareous podzolized light loam soil on a "rikkh" carbonaceous moraine. Koeru, Estonian Republic.

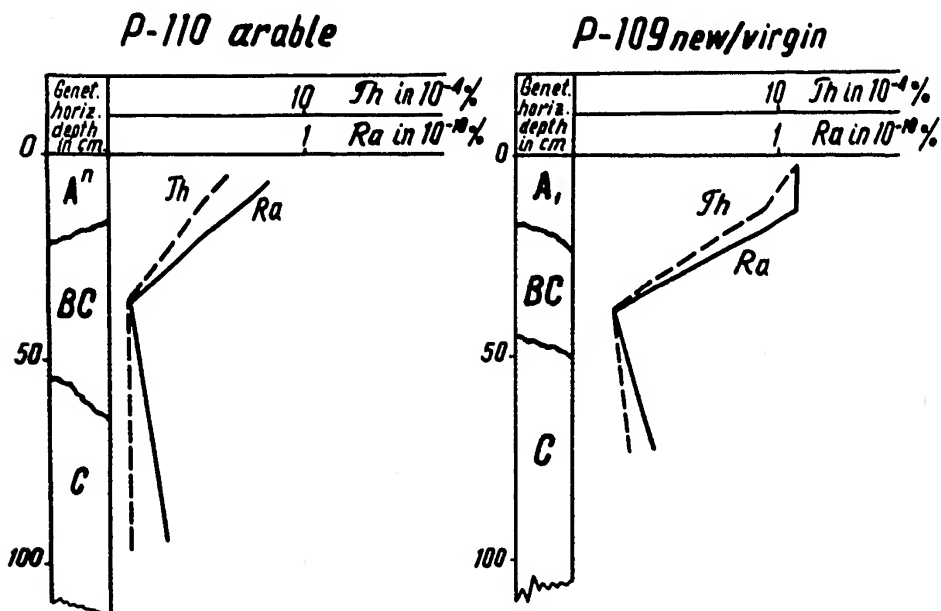


Fig. 4 Distribution of Ra and Th in soddy-calcareous typical thin sandy loam soil on fluvio-glacial deposits. Imperi, Estonian Republic.

Fig. 5 Distribution of Ra and Th in humus-calcareous light loamy soil with limestone bedding. State farm Aseri, Estonian Republic.

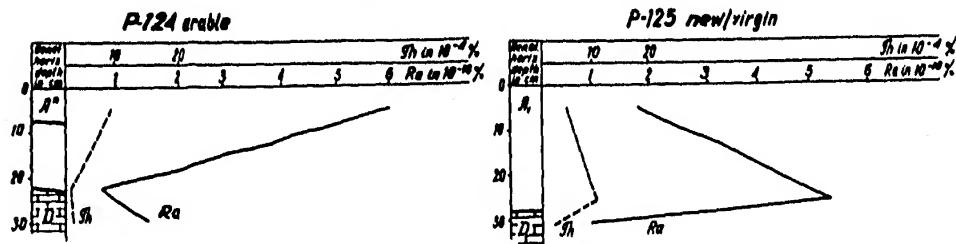


Fig. 6 Distribution of strontium-90 in the surface horizon of the soil covering in the USSR (0-5 cm) by latitudinal zones (the middle of 1960). mc/km^2

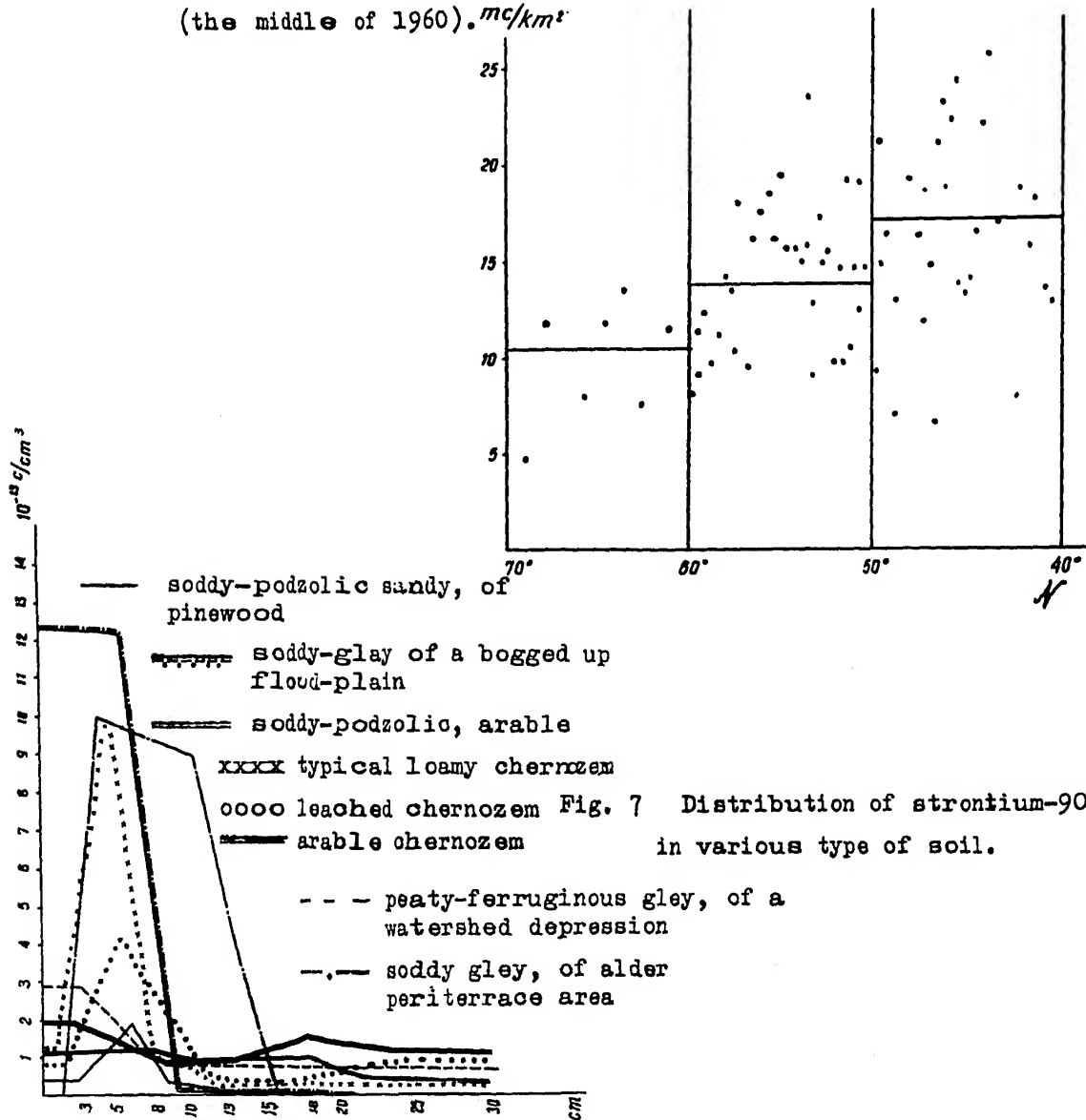


Fig. 7 Distribution of strontium-90 in various type of soil.